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THE EFFECT OF IODINE ON THE MATERIAL  
PROPERTIES OF PACKAGING POLYMERS

Frederick J. Dechow, et al

Army Materials and Mechanics Research Center  
Watertown, Massachusetts

December 1973

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# THE EFFECT OF IODINE ON THE MATERIAL PROPERTIES OF PACKAGING POLYMERS

FREDERICK J. DECHOW and NATHANIEL S. SCHNEIDER  
POLYMER & CHEMISTRY DIVISION

December 1973



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ABSTRACT

Films of nine polymers were examined to determine any changes in their physical and mechanical properties after exposure to solutions of iodine and potassium iodide. The sorption and permeability of iodine for the films were measured at temperatures of 105 F and 140 F. These films were also analyzed by X-ray diffraction and by thermomechanical studies. The results are discussed in terms of differences in polymeric repeat units and in terms of crystallinity. Recommendations for future studies are included. (Authors)

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## PREFACE

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## INTRODUCTION

The plastic strip packaging of water purification tablets offers many advantages over bottling of these tablets. However, significant problems have been encountered in developing strip packaging and samples produced in pilot production runs have not provided long-term tablet stability which is required.

The iodine water purification tablet loses its effectiveness when exposed to moisture or when the iodine content is reduced by sublimation. In the heat-sealed strip package, each tablet is enclosed in an individual thermoformed bubble. The tablet so packaged is required to maintain its integrity under conditions of 100 F, 95% relative humidity (RH) for a period of two or more years in storage. The success of this approach depends on the selection of a polymer film which is impermeable to moisture, resistant to iodine vapor, and readily heat sealable. At present, satisfactory packaging has not been achieved and failures occur within two months under the above specified test conditions. This situation has motivated the present study to obtain knowledge of iodine and water interactions with polymers of different structure to assist in a successful materials engineering solution to this problem.

Nine different polymer packaging films were selected for testing at this laboratory. The repeat units of these polymers are listed in Table 1 along with the thickness of the samples used. The polypropylene was purchased from Phillips Petroleum Company. Some samples were hot stretched to observe the effect of this treatment on its material properties. The sample of Lexan (polycarbonate) a trademarked product of General Electric was obtained from Rowland Products. The Teflon (polytetrafluoroethylene) and the Mylar [poly(ethylene terephthalate)] which are trademarks owned by the DuPont Company and the polyethylene were purchased from Blue Bell Plastics. The Aclar, produced by Allied Chemical Corporation, came in three forms: 22A, a copolymer of 96% chlorotrifluoroethylene and 4% vinylidene fluoride with approximately 34% crystallinity; 22C, a copolymer like 22A but with approximately 55% crystallinity; 33C, a terpolymer of 96% chlorotrifluoroethylene, 1% vinylidene fluoride, and 3% tetrafluoroethylene with approximately 55% crystallinity. The Saranex (coextruded polyethylene and polyvinylidene chloride) and PZ2000.36, a similar coextruded polymer film, were samples from Dow Chemical. These polymers were selected because they are used in packaging and because they are expected to provide a range of behavior from highly active to highly inert with respect to iodine.

The rate of water vapor transport at 95% RH and 100 F for these polymers are listed in Table 2 with the literature references where the values were obtained. Two values are listed for polycarbonate (Lexan) because of the large difference in the transport rate given by the two sources.

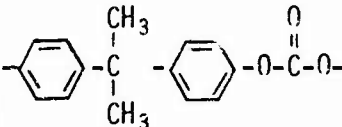
## SORPTION STUDIES

### Experimental

The sorption experiments were carried out to determine the amount of iodine uptake for the polymer samples at different temperatures and for a series of



Table 1. TEST POLYMER FILMS

Material	Repeat Unit	Thick- ness (mils)	Crystal- linity
Polypropylene	$-\text{CH}_2-\text{CH}-$ $\text{CH}_3$	2.5	Moderate
Lexan:Polycarbonate		2.0	Low
Teflon:Polytetrafluoroethylene	$-\text{CF}_2-\text{CF}_2-$	1.5	High
Mylar:Poly(ethylene terephthalate)	$\text{CH}_2-\text{CH}_2-\text{O}-\overset{\text{O}}{\parallel}\text{C}-\text{C}_6\text{H}_4-\overset{\text{O}}{\parallel}\text{C}-\text{O}-$	3.0	Moderate
Polyethylene	$-\text{CH}_2-\text{CH}_2-$	2.0	High
Aclar 22 (chlorotrifluoroethylene 96% : (vinylidene fluoride 4%	$-\text{CFCl}-\text{CF}_2-$ $-\text{CH}_2-\text{CF}_2-$	3.0	Moderately high
Aclar 33 (chlorotrifluoroethylene 96% (vinylidene fluoride 1% (tetrafluoroethylene 3%	$-\text{CFCl}-\text{CH}_2-$ $-\text{CH}_2-\text{CF}_2-$ $-\text{CF}_2-\text{CH}_2-$	3.0	Moderately high
Saranex Polyethylene Poly(vinylidene chloride) Polyethylene	$-\text{CH}_2-\text{CH}_2-$ $-\text{CH}_2-\text{CCl}_2-$ $-\text{CH}_2-\text{CH}_2-$	1.2	Moderate
PZ2000.36 Polyethylene Poly(vinylidene chloride) Polyethylene	$-\text{CH}_2-\text{CH}_2-$ $-\text{CH}_2-\text{CCl}_2-$ $-\text{CH}_2-\text{CH}_2-$	2.0	Moderate

iodine concentrations. Initially, these experiments were performed using the improved test cell developed by Rogers, Pillion, and Kaplan\* for measuring the performance of packaging materials against chlorine and iodine. The cell consists of two dishes made from 60-mm Pyrex standard wall tubing, flanged and ground so that a tight seal is formed when one dish is inverted on top of the other. Two small glass hooks are fused in the top dish near its center. Whereas Rogers et al. used the hooks to hold a beaker with their indicator solution, in the current experiments they were used to suspend a polymer test film. A second sample of the polymer test film was placed in the 25 ml of iodide solution in the bottom dish. Thus, with the dishes sealed with a silicone lubricant, one sample was

\*ROGERS, M. R., PILLION, E., and KAPLAN, A. M. *Modern Packaging*, November 1957.

soaking in the iodide solution and the second sample was exposed to its vapors. The test cells were placed for two weeks in an Aminco environmental chamber maintained at constant temperature.

Later, the experiments were performed using 250-ml and 125-ml Erlenmeyer flasks with ground glass stoppers. In these experiments, three samples of a polymer were placed in the flasks with either 225-ml or 100-ml of the iodine solution. The flasks were then placed in the Aminco environmental chamber for two weeks. This change was made in the procedure because the weight changes for some of the polymers samples using Rogers' cells were so slight as to be inconclusive.

For both sets of experiments, blanks were run using distilled water in place of the iodine solution. The water was not sorbed significantly (<1%) by any of the polymer films. Therefore, weight changes in the polymer samples after exposure to the iodine solutions are interpreted as the sorption of iodine by the polymer film.

#### Results

The temperature of the Aminco environmental chamber was set at 105 F for one series of experiments (Table 3) and at 140 F for the second series (Table 4). The iodine concentrations for 105 F were  $0.05\text{MI}_3$ ,  $0.25\text{MI}_3$ , and  $1.00\text{MI}_3$ . For 140 F, they were  $0.25\text{MI}_3$ ,  $0.50\text{MI}_3$ , and  $1.00\text{MI}_3$ .

Table 2. WATER VAPOR TRANSPORT

Material	Rate of Water Vapor Transport $\frac{(g)(mil)}{(m^2)(24 \text{ hr})}$	Reference
Polypropylene	72	TAPPI, v. 44, 1961, p. 58
Lexan	232 1983	Mod. Plastics Encycl., 1972-1973 J. Appl. Polymer Sci., v. 7, 1963, p. 1649
Teflon	4.8	Ind. Eng. Chem., v. 45, 1953, p. 2296
Mylar	38	Ind. Eng. Chem., v. 45, 1953, p. 2290
Polyethylene	17	Ind. Eng. Chem., v. 45, 1953, p. 2290
Aclar 22A	0.70	Allied Chemical Technical Data
Aclar 22C	0.87	Allied Chemical Technical Data
Aclar 33C	0.46	Allied Chemical Technical Data
Saranex	6.14	Dow Chemical Company Technical Data
PZ2000.36	6.98	Dow Chemical Company Technical Data

Table 3. SORPTION OF IODINE BY THE POLYMER FILMS AT T=105 F

Material	$0.05\text{MI}_3$			$0.25\text{MI}_3$			$1.0\text{MI}_3$		
	Weight			Weight			Weight		
	Weight (g)	Change (g)	(%) Change	Weight (g)	Change (g)	(%) Change	Weight (g)	Change (g)	(%) Change
Saranex	0.0703	0.0098	13.94	0.0375	0.0061	16.27	0.0656	0.0220	33.5
PZ2000.36	.1050	.0055	5.24	.1465	.0106	7.23	.1714	.0260	15.17
Polyethylene	.1058	.0002	0.76	.1000	.0010	1.00	.0454	.0008	1.76
Polycarbonate	.0556	.0020	3.60	.1471	.0056	3.81	.1350	.0200	14.81
Teflon	.1172	.0010	0.85	.1464	.0006	0.41	.859	.0011	0.59
Mylar	.2347	.0017	0.72	.2148	.0021	0.98	.0663	.0008	1.20
Aclar 22C	.2762	.0004	0.14	.4976	.0000	0.00	.4585	.0002	0.04
Aclar 22A	.2584	.0004	0.15	.5989	.0002	0.03	.5027	.0005	0.10
Aclar 33C	.3282	.0001	0.03	.3275	.0002	0.06	.4659	.0001	0.02

Table 4. SORPTION OF IODINE BY THE POLYMER FILMS AT T=140 F

Material	$0.25\text{MI}_3$			$0.50\text{MI}_3$			$1.0\text{MI}_3$		
	Weight			Weight			Weight		
	Weight (g)	Change (g)	(%) Change	Weight (g)	Change (g)	(%) Change	Weight (g)	Change (g)	(%) Change
Saranex	0.3510	1.6119	459.2	0.3730	2.1412	574.0	0.4039	1.7043	487.7
PZ2000.36	0.9282	1.2587	135.6	1.1037	1.6029	145.23	1.0896	2.9586	271.53
Polyethylene	0.5877	0.0276	4.6	0.6630	0.0454	6.84	0.5019	0.0264	5.26
Polycarbonate	0.5564	*	*	0.5184	*	*	0.6356	*	*
Polypropylene	0.4074	0.0011	0.298	0.3695	0.0020	0.49	0.3842	0.0030	0.78
Teflon	0.9331	0.0007	0.075	0.8640	0.0001	0.012	0.5998	0.0005	0.08
Mylar	0.9906	0.0549	5.542	0.9476	0.0551	5.819	1.2303	0.1615	13.128
Aclar 22A	2.1074	0.0007	0.033	2.8031	0.0027	0.096	2.0069	0.0004	0.019
Aclar 33C	1.8891	0.0007	0.37	2.5046	0.0022	0.080	2.9154	0.0030	0.103

\*These samples became brittle and disintegrated in their removal from the flasks.

The Saranex films were blackened by their exposures to iodine at all concentration levels. While the films were smooth initially and at the end of the 105 F experiment, for 140 F the films were rough and bumpy. Saranex showed the greatest absorption of iodine - almost six times its own weight in one instance. There seems to be a peak sorption near  $0.5\text{MI}_3$  because an increase in iodine concentration does not increase the amount sorbed.

The PZ2000.36 films were likewise blackened by their exposure to iodine and became rough at the 140 F experiments. There is no decrease in the amount of iodine sorbed after the  $0.5\text{MI}_3$  concentration; rather, the amount sorbed continues to increase with concentration increases through the experiments. While the amount of iodine sorbed is not as large a percent change in sample weight as for Saranex, for the 140 F experiments the amount sorbed is more than the polymer weight.

Polyethylene blackens on exposure to iodine but sorbs very little iodine compared to the coextruded films of polyethylene and polyvinylidene chloride. In both these multilayer films it appears that the inner Saran layer separated from the polyethylene outer layers and that iodine solution accumulated in the blistered areas, thus accounting for the exceptionally high apparent sorption levels. One should note that it exhibits the same kind of maximum in sorption as Saranex near  $0.5\text{MI}_3$  at 140 F.

Polycarbonate turns a pale pink when exposed to the iodine solutions at 105 F. At 140 F the polymer film is a brownish grey for  $0.25\text{MI}_3$ . This color progresses to a deep black for  $1.0\text{MI}_3$ . All of the samples at 140 F became very brittle and shattered during their extraction from the flasks. Therefore, there are no weight changes recorded.

Polypropylene, although it sorbed very little of the iodine, discolored to a light red at all concentrations. When polypropylene is hot stretched, it does not discolor when exposed to iodine solutions.

Teflon showed no color change and no sorption of iodine at any concentration or at any temperature.

Mylar discolored upon exposure to iodine and showed significant sorption for the iodine solutions at 140 F. The color ranged from a light red for the experiments at 105 F to a deep black for  $1.0\text{MI}_3$  at 140 F.

None of the Aclar samples discolored for the 105 F experiments but they did become a very light pink for the iodine solutions at 140 F. No significant weight change accompanied this slight discoloration.

For those polymers that did show measurable iodine uptake in the Rogers' type test cell the same percentage increase was noted for the sample in the vapor as for the sample in solution.

## PERMEABILITY STUDIES

### Experimental

The permeability experiments were carried out to determine the ease with which iodine passes through the polymer samples at different temperatures. These experiments were performed using the test cells described in the Sorption Studies section. In these experiments, however, the polymer films separated the upper and lower dishes. From the hooks in the top dish was suspended a 1-ml beaker filled with a water solution of Thyodene (Fisher), a reagent indicator for iodine. The test cells were placed in an Aminco environmental chamber maintained at constant temperature. In the lower dish was 25 ml of 0.1M  $I_3^-$  solution. The vapors of the iodine solution may diffuse through the polymer film to the indicator solution. The indicator solutions were checked hourly for the first eight hours and then daily for the next thirty days.

### Results

The temperature of the Aminco environmental chamber was set at 105 F for one set of experiments and at 140 F for the second set. Two different phenomena have been noted in these experiments: the discoloration of the polymer film and the change in the indicator solution. The results for the ambient temperature equal to 105 F are recorded in Table 5. Those for 140 F are recorded in Table 6. Permeability experiments with hot-stretched polypropylene indicated just as rapid indicator change as unstretched polypropylene, although there was no discoloration of hot-stretched film.

## X-RAY DIFFRACTION STUDIES

Wide angle X-ray diffraction photographs were made to determine if the crystalline structure of the polymer samples had been altered by the exposure to the iodine solutions. A Warhaus diffraction camera used the  $CuK\alpha$  radiation collimated by a pinhole of 0.2-mm diameter to scatter from the polymer sample at the 0.15-mm diameter receiving slit. The photographic film was usually placed 5.35 cm from the sample.

Typical photographs are shown in Figure 1 for polyethylene and Figure 2 for polypropylene samples. The diffraction patterns 1a and 2a are from samples that had been exposed to water for two weeks at 105 F. The diffraction patterns 1b and 2b are from samples exposed to 1.0M  $I_3^-$  for two weeks at 140 F. There appears the expected evidence of crystalline orientation for the polypropylene and the polyethylene. Diffraction photographs after the iodine treatment were taken with the film 6.50 cm from the sample to increase the resolution. However, there appear to be no detectable changes in either of these cases due to the exposure to the iodine solutions.

Similar diffraction pictures gave the expected crystalline pattern for Teflon and Mylar and amorphous patterns for polycarbonate. Again, no changes were noticed between photographs taken prior to and after the two-week exposure to the iodine solutions suggesting that in the cases studied here the iodine was sorbed into amorphous regions of the polymer leaving the crystalline structure unaffected.

# THERMOMECHANICAL STUDIES

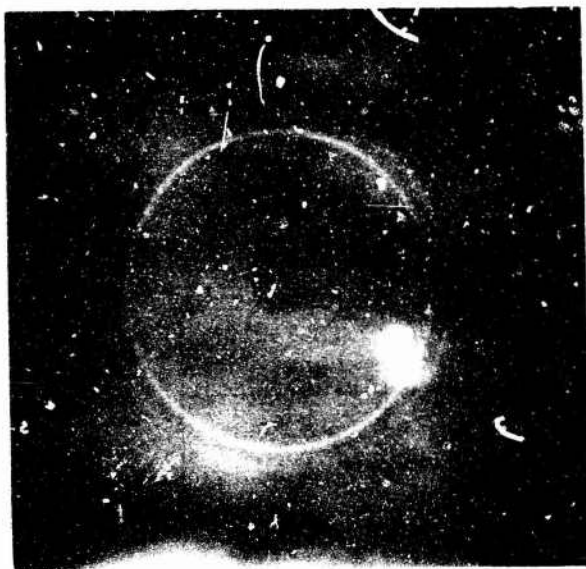
A Rheovibron DDV-II instrument has been used at a frequency of 110 Hz to study the dynamic mechanical properties of the polymer samples as a function of temperature. This study was done on the polymers before and after the two weeks at 140 F in the iodine solutions. Figure 3 shows results for Saranex.

Table 5. PERMEATION RESULTS FOR T=105 F

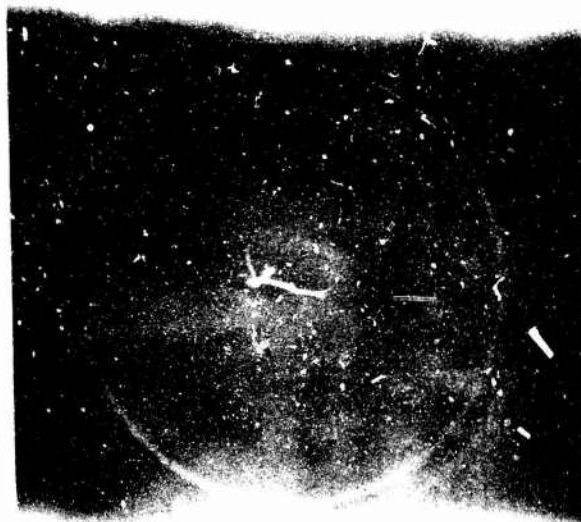
Material	Film Thickness(mils)	Discoloration Time	Indicator Change Time
Polyethylene	2.0	3 hours	8 hours
Polypropylene	2.5	5 hours	8 hours
Polycarbonate	2.0	8 days	11 days
Saranex	1.2	4 hours	1 day
PZ2000.36	3.0	6 hours	3 days
Teflon	1.5	none after 30 days	3 days
Mylar	3.0	11 days	none after 30 days
Aclar 22A	3.0	none after 30 days	none after 30 days

Table 6. PERMEATION RESULTS FOR T=140 F

Material	Film Thickness(mils)	Discoloration Time	Indicator Change Time
Polyethylene	2.0	2 hours	5 hours
Polypropylene	2.5	4 hours	5 hours
Polycarbonate	2.0	3 days	4 days
Saranex	1.2	2 hours	6 hours
PZ2000.36	2.0	3 hours	6 hours
Teflon	1.5	none after 30 days	2 days
Mylar	3.0	6 days	none after 30 days
Aclar 22A	3.0	none after 30 days	none after 30 days

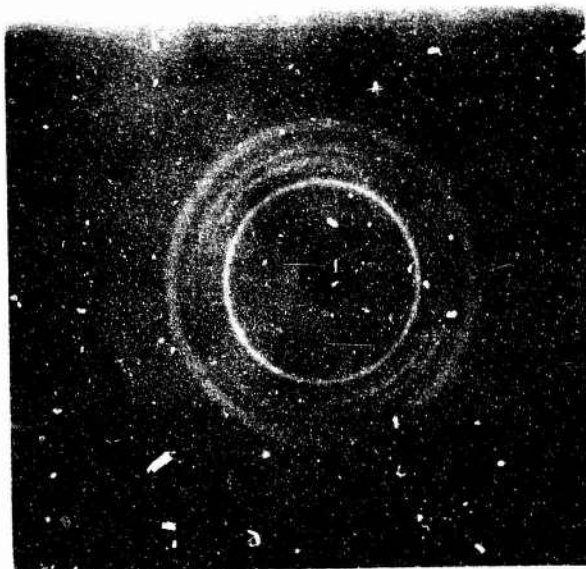


a. Without Iodine Treatment (5.35 cm)

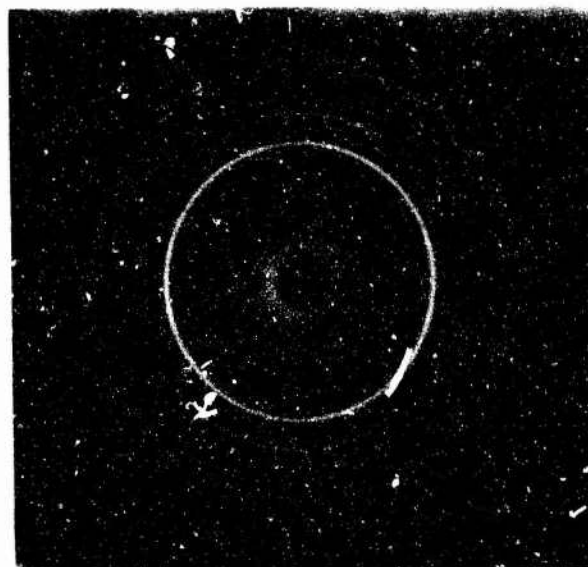


b. After Two Weeks in a  $1.0\text{M I}_3^-$  Solution. (6.50 cm)

Figure 1. Diffraction Patterns Taken of Samples of Polyethylene



a. Without Iodine Treatment (5.35 cm)



b. After Two Weeks in a  $1.0\text{M I}_3^-$  Solution. (6.50 cm)

Figure 2. Diffraction Patterns Taken of Samples of Polypropylene

One should note that the low temperature transition for the polyethylene in this coextruded sample is distinctly present both before and after the iodine treatment. However, the real part of the dynamic modulus,  $E'$ , has a much more pronounced drop at  $-25^\circ\text{C}$ . Also the values for  $E'$  are lower in Figure 3b.

The differences between the exposed and nonexposed samples for the polymer films of polyethylene, polypropylene, and Teflon were within the experimental uncertainty of the instrument. This suggests that the sorbed iodine does not result in any significant alteration of the structure of these three polymers and is not involved in specific chemical interactions with the polymer groups.

## DISCUSSION

The effect of the different polymeric structures on the amount of iodine sorbed can be readily seen. Polyethylene and polypropylene are both moderately crystalline and differ only in a pendant methyl group in the repeat unit. Yet

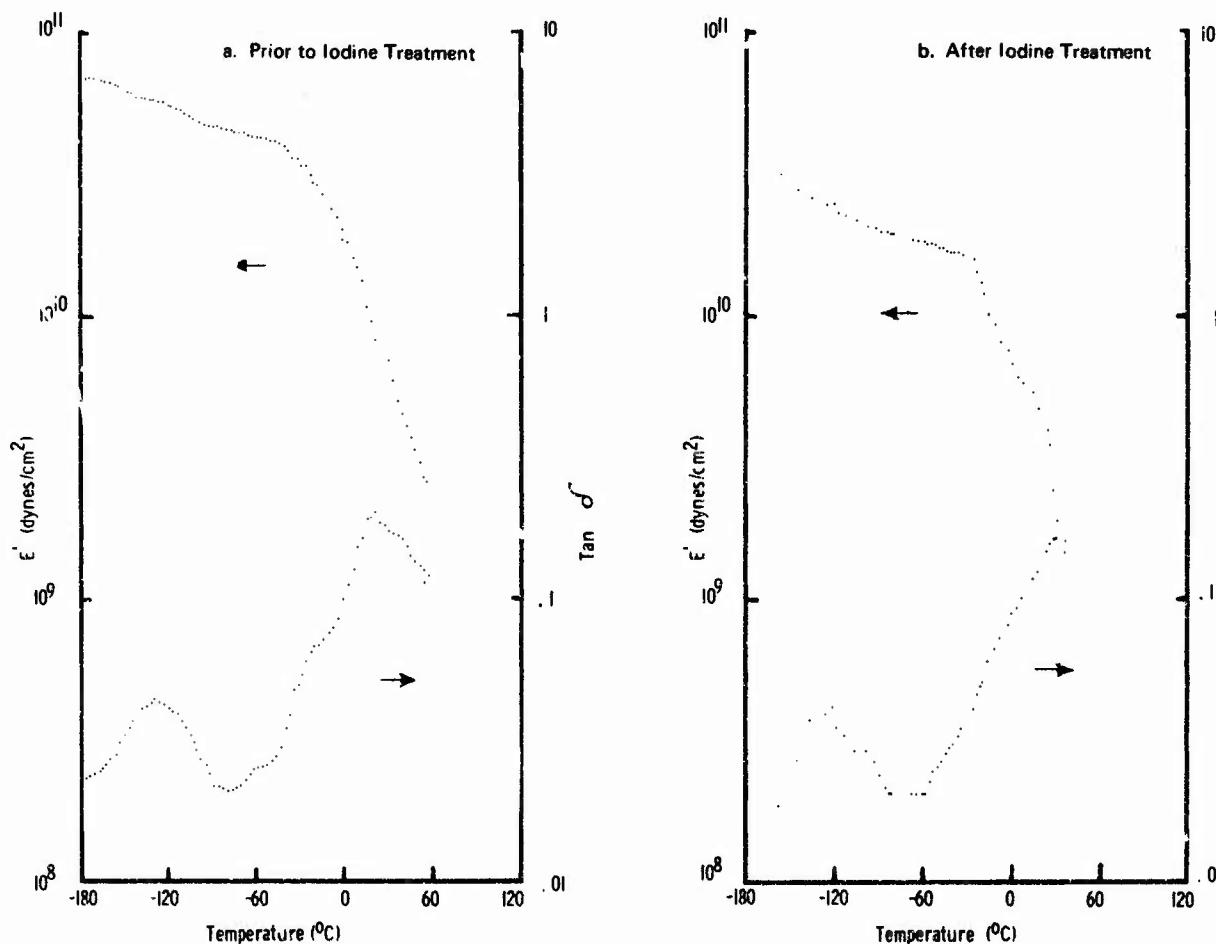


Figure 3. Dynamic Modulus of Saranex



polyethylene absorbs enough iodine to blacken (about 5% of the sample weight for a 1.0M I<sub>2</sub> solution at 140 F) while the polypropylene becomes only light red under the most extreme conditions and does not absorb a significant amount of iodine. However, neither material is even a moderate barrier to iodine, it being a matter of hours for the iodine to permeate them.

The two polyesters, Mylar and polycarbonate, resist sorption of iodine for a time. Once the discoloration process begins, polycarbonate loses its material integrity and allows iodine permeation. Under extreme conditions, polycarbonate becomes so structurally weakened as to shatter when it is touched. Mylar, however, absorbs very moderate amounts of iodine and does not allow iodine to permeate within thirty days. The repeat units of these two polyesters are essentially the same, the only structural distinction is the low crystallinity of polycarbonate compared to moderate crystallinity of Mylar. This would not explain the large observed differences in stability which in this case are the results of chemical and probably oxidative interactions.

Teflon has the same backbone as polyethylene with fluorine in place of the hydrogen atoms. The Teflon polymer samples are highly crystalline and fibrillous which allows for the relatively easy passage of iodine through it. However, there is no sorption of iodine by Teflon.

Saranex and PZ2000.36 interact strongly with iodine in their inner poly(vinylidene chloride) layers. This has been confirmed by viewing a cross section of the film with an optical microscope. Under the microscope one can see the reaction pockets in the center of the sample which are the roughness one observes without the microscope. Further, one can pull gently on a small section of these polymer samples which have been in the iodine solutions to separate the polyethylene layers from the polyvinylidene chloride layers. The polyethylene in the coextruded polymer film appears to react to the same extent as when it is alone. This should be investigated further to determine if it is true.

The copolymer and terpolymers of Aclar are moderately high in crystallinity and sorb only enough to slightly discolor it in the most severe conditions. Although the Aclar samples discolored very slightly in the iodine solutions at 140 F, in the permeability experiments when samples were exposed to the iodine vapor alone at that temperature there was no discoloration. These points indicate the requirement for a fluorine-substituted ethylene-backbone polymer for maximum impermeability to iodine. While moderate to high crystallinity appears to be needed for most materials to be impervious to iodine sorption, Aclar 22A with only 35% crystallinity still sorbs an insignificant amount.

## CONCLUSIONS AND RECOMMENDATIONS

Several general statements can be made regarding the polymer iodine interactions. The results indicate that iodine is appreciably soluble in several of the polymers studied and transmission rates can be high enough that the loss of iodine would affect the titer of the purification tablet in a relatively brief span of time. The iodine sorption process in the polymers studied appears to be physical in nature; there is little evidence for specific chemical interactions

which alter the structure of the polymers or lead to complexation with specific groups. On the other hand, only two of the eight polymers studied, Lexan and Mylar, have polar functional groups and in polyacrylonitrile specific interactions with the nitrile groups have been demonstrated.

The marked difference in iodine sorption levels, permeability and stability for Lexan and Mylar is puzzling in view of apparent similarities in the molecular structure of these two polymers. These differences can be rationalized only partially in terms of differences in crystallinity but are not peculiar to iodine since the water transmission rate is fifty times greater in Lexan than in Mylar (Table 2).

In substance, the present work confirms the earlier conclusion that Mylar and Aclar are the two materials most resistant to iodine permeability. If, as it appears, moisture transmission rates are also important, then the Aclar materials offer a clear advantage over Mylar for the strip packaging of the iodine water purification tablets.

There are several approaches which might be pursued to develop suitable strip packaging. The problem of forming a good seal has been discussed with Allied Chemical which produces Aclar film. Allied Central Research Laboratories has indicated that it has developed effective production sealing methods different from those used in prior test production runs on tablet packaging which may warrant further investigation.

A second engineering approach involves the use of bilayer materials with an inner layer, such as polyethylene, to facilitate the sealing process. Even if the polymer used for the seal is relatively permeable to iodine or moisture, an effective barrier can be formed due to the long path length for diffusion represented by the width of the seal. Bilayer fluorohalocarbon films with polyvinylchloride and with polyethylene are available and have been used successfully for packaging pharmaceuticals of extreme moisture sensitivity.

If the above engineering solutions prove to be ineffective, then further research directed toward the selection or development of new polymers would be warranted. For example, the Japanese have announced an ethylene naphthenate film which has one third the moisture transmission rate of Mylar and is clear. If this film can be readily sealed, it might provide a useful and less expensive alternative to Aclar. There are probably other commercial polymers worth screening including those not normally provided in film form such as polyaryl ethers, polysulfone, chlorinated polyether, etc. If any synthetic research is deemed necessary for the development of new polymers, then the present study suggests that highly fluorinated materials with moderate crystallinity would offer the most promise. A number of interesting polymers of this type have been prepared by the National Bureau of Standards. However, it must be pointed out that the high cost of any such specialty polymers would probably render this last approach impractical.